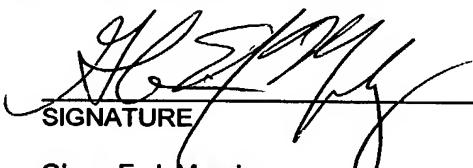


Form PTO-1390 <small>(Rev. 5-93)</small> TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NO. H 4136 PCT/US
		<small>U.S. APPLICATION NO. (if known; see 37 CFR 1.5)</small> 107009460
INTERNATIONAL APPLICATION NO. PCT/EP00/03852	INTERNATIONAL FILING DATE April 28, 2000	PRIORITY DATE CLAIMED May 7, 1999
TITLE OF INVENTION THERMOPLASTIC COMPOSITE MATERIAL		
APPLICANT(S) FOR DO/EO/US Juergen LORENZ and Ewald WILKA		
<p>Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:</p> <ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). <p>Items 11. to 16. below concern other document(s) or information included:</p> <ol style="list-style-type: none"> 11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 14. <input type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input checked="" type="checkbox"/> Other items or information.: International Search Report (With Information Disclosure Citation and References) <p>"Express Mail" mailing label number <u>EL 843287890 US</u></p>		

U.S. Application No. (If known see CFR1.30)	INTERNATIONAL APPLICATION NO. PCT/EP00/03852	ATTORNEY'S DOCKET NUMBER H 4136 PCT/US		
10/009460				
<p>17. ■ The following fees are submitted:</p> <p>Basic National Fee (37 CFR 1.492(a)(1)-(6)): Search Report has been prepared by the EPO or JPO..... \$890.00</p> <p>International preliminary examination fee paid to USPTO (37CFR 1.482) \$670.00</p> <p>No International preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37CFR 1.445(a)(2))..... \$690.00</p> <p>Neither International preliminary examination fee (37CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$970.00</p> <p>International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$96.00</p>		CALCULATIONS PTO USE ONLY		
ENTER APPROPRIATE BASIC FEE AMOUNT		= \$ 890 00		
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).		\$ 0 00		
Claims	Number filed	Number Extra	Rate	
Total Claims	13 - 20 =	0	0 X \$18.00	\$ 0 00
Independent Claims	2 - 3 =	0	0 X \$78.00	\$ 0 00
Multiple dependent claims (s)(if applicable)		0	+ \$260.00	\$ 0 00
TOTAL OF ABOVE CALCULATIONS		=	\$ 890	00
Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$ 0	00	
SUBTOTAL		=	\$ 890	00
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37CFR 1.492(f)).		\$ 0	00	
		+		
TOTAL NATIONAL FEE		=	\$ 890	00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$ 0	00	
		+		
TOTAL FEES ENCLOSED		=	\$ 890	00
		Amount to be: refunded	\$-----	
		charged	\$890.00	
<p>a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.</p> <p>b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>01-1250</u> in the amount of <u>\$890.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>01-0789</u>.</p> <p>c. <input checked="" type="checkbox"/> The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>01-1250</u>. A triplicate copy of this sheet is enclosed.</p> <p>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.496 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</p>				
<p>SEND ALL CORRESPONDENCE TO: Henkel Corporation, Law Dept. 2500 Renaissance Blvd, Suite 200 Gulph Mills, PA 19406</p> <p> SIGNATURE</p> <p>Glenn E. J. Murphy NAME ATTORNEY FOR APPLICANT 33,539 REGISTRATION NUMBER</p>				

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PATENT
Docket H 4136 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Re: PCT/EP00/03852

International Filing Date: April 28, 2000

Priority Date: May 7, 1999

Applicant: LORENZ, et al.

Title: THERMOPLASTIC COMPOSITE MATERIAL

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents
Washington, DC 20231

Please enter the amendments below before examining this case on the merits:

IN THE SPECIFICATION:

On page 1, insert below the title:

--CROSS-REFERENCE TO RELATED APPLICATIONS

This application is A U.S. National Stage application filed under 35 U.S.C. § 371 of International Application No. PCT/EP00/03852, filed April 28, 2000, in the European Patent Office, claiming priority under 35 U.S.C. §§ 119 and 365 of PCT/EP00/03852 and DE 199 21 209.0, filed on May 7, 1999, in the German Patent Office.--

On page 4, between lines 1 and 2, insert the heading -- DESCRIPTION OF THE INVENTION--.

Preliminary Amendment of US National Stage for International Application PCT/EP00/03852 filed April 28, 2000

IN THE CLAIMS:

Please cancel claims 1 to 13 without prejudice, and add new claims 14 - 26:

14. A thermoplastic composite material comprising:

- a) at least 15% by weight of an organic fibrous material or of a mixture of two or more organic fibrous materials; and
- b) at least 15% by weight of a thermoplastic binder;

wherein the binder comprises at least 10% by weight of one or more polymers selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates, ethylene/vinyl acetate copolymers, and copolymers of two or more of the polymers, wherein the binder does not consist solely of two different polyacrylates.

15. The composite material of claim 14, comprising 15% to 45% by weight of the organic fibrous material.

16. The composite material of claim 14, comprising at least 40% by weight of the thermoplastic binder.

17. The composite material of claim 15, comprising at least 40% by weight of the thermoplastic binder.

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18. The composite material of claim 14, wherein thermoplastic binder comprises at least 30% by weight of polyvinyl acetate.

19. The composite material of claim 14, wherein the thermoplastic binder comprises at least 10% by weight of a copolymer of butadiene and styrene.

20. The composite material of claim 14, further comprising up to 20% by weight of one or more ingredients selected from the group consisting of inorganic salts, cationic polymers, preservatives, dyes, natural fats, synthetic fats, paraffins, natural oils, synthetic oils, silicone oils, ionic surfactants, and nonionic surfactants.

21. The composite of claim 14, wherein the organic fibrous material comprises one or more synthetic fibers, vegetable fibers, or animal fibers.

22. The composite material of claim 14, wherein the organic fibrous material comprises leather fibers.

23. The composite material of claim 14, wherein the organic fibrous material comprises fibers having a stretched length of about 0.1 mm to about 15 mm.

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24. A process for the production of a thermoplastic composite material, comprising the steps of:

- a) combining in any order and mixing one or more organic fibrous materials comprising fibers having a stretched fiber length of 0.1 mm to 15 mm with one or more polymer dispersions, each dispersion comprising one or more polymers selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl acetate copolymers, and copolymers of two or more of the polymers, wherein the polymers comprising the dispersion or dispersions in combination do not consist solely of two different polyacrylates;
- b) treating the resultant mixture with an aqueous solution of an aluminum salt or copper salt or mixture thereof; and
- c) dewatering and drying the treated mixture from step b), to form a thermoplastic composite material comprising at least 15% by weight of the one or more organic fibrous materials and at least 15% by weight of the one or more polymers as thermoplastic binder.

25. An article of manufacture comprising the thermoplastic composite material of claim 14, laminated with a polymer film on a substrate surface.

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26. A process surface coating of a substrate, comprising the steps of bonding the thermoplastic composite material of claim 14 to a substrate surface with a hotmelt adhesive.

REMARKS

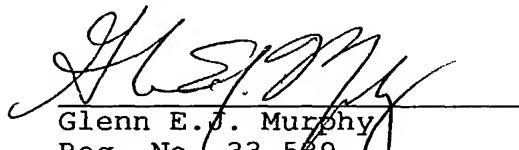
Applicants cancel claims 1-13 without prejudice and enter claims 14-26. The subject matter of the new claims is described in the original claims. The specification has been amended to include a cross-reference to related applications and headings appropriate to U.S. practice. No new matter has been added.

The new claims better claim the full literal and equivalent scope and breadth of subject matter disclosed in the application, notwithstanding applicants' belief that the original claims, drafted for examination in the German and European Patent Offices, would have been allowable but for minor matters of form permitted in German or European practice but objected to in the U.S.P.T.O. The new claims find support in the application independent of the original claims and therefore are not believed to constitute narrowing amendments to the original claims within the holding of Festo Corp. v. Shoketsu Kinzoku Kogyo Kabushiki Co., No. 95-1066 (Fed. Cir. Nov. 29, 2000).

Preliminary Amendment of US National Stage for International
Application PCT/EP00/03852 filed April 28, 2000

Applicants respectfully request entry of this Amendment
and examination of the application. If any fees are due to
enter this paper that have not been accounted for, please
charge Deposit Account No. 01-1250.

Respectfully submitted,



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Thermoplastic Composite Material

This invention relates to a thermoplastic composite material containing an organic fibrous material or a mixture of two or more organic fibrous materials and a thermoplastic binder. The invention also relates to a process for producing the thermoplastic composite material and to the 5 use of the composite material for coating surfaces of objects and to the objects thus coated. The invention also relates to an adhesive for bonding the composite material to the object to be coated.

Composite materials are materials which are formed by incorporating a base material, for example in the form of fibers, in a second 10 material (the matrix). Certain properties (for example mechanical properties, surface properties or certain behavior towards external influences) of the incorporated material are utilized for the composite material. The base material may vary widely in its quantity ratio to the matrix surrounding it. Thus, the percentage matrix content of wood-based 15 materials, which include for example the known presspahn boards, is generally only 10 to 15%. By contrast, the percentage matrix content of fiber-reinforced plastics, for example glass fiber-reinforced plastics, can be significantly higher, for example more than 70% or even more than 80%.

In many cases, it is possible by suitably selecting the base material 20 and the matrix to impart to the composite material certain properties of the base material which are paired with certain properties of the matrix. For example, the use of glass or natural fibers in thermosets can cause the tensile strength of the fibers to be transferred to the plastic matrix which, in turn, contributes further advantages to the composite material in regard to 25 shaping, dimensional stability and processability.

In many cases, the object of making composite materials is to produce from by-products accumulating in the processing of a certain base

- material a material which possesses characteristic properties of that base material. The corresponding composite material may then generally be used at least as a substitute for the base material and thus provides for the "substance-related" utilization of the base material waste or by-products.
- 5 This is the case, for example, with the utilization of wood waste in presspahn boards. Another example of the substance-related utilization of waste products is the processing of cutting and stamping waste from the manufacture of leather and shoes to leather fiber materials.
- Leather waste can be reduced to fibers and can then be processed
10 to leather fiber materials (LEFAs). LEFAs are generally single-layer materials of leather fibers and binders. Even at the end of the thirties, LEFA boards were being used in the shoe industry, for example for the production of counters, insoles and inner soles, slipper soles, heels and welts. LEFA materials were also used to a small extent for industrial
15 leather seals.
- Besides such properties as flexibility and durability which qualify leather for use in the shoe industry and in the leather goods industry, leather is being increasingly used in other areas where the decorative features of leather are predominant. However, it has hitherto been difficult
20 or even impossible to use leather as a surface material for furniture fronts, wall and ceiling panels or other consumer goods. In particular, it was particularly difficult to use leather as a surface material for objects with a three-dimensional relief structure.
- Since leather does not have any thermoplastic properties, it can only
25 be used to a limited extent as a surface coating material. In particular, it is difficult to provide varying three-dimensional relief structures with a smooth leather surface by an economical process.
- Such economical processes for coating relief-like surfaces of moldings are known, for example, from the film coating industry. In this
30 case, a molding (object) is generally coated with a thermoplastic film by

heating the molding and the film to a temperature above the flow transition limit of the plastic film and suitably drawing the film (for example by applying a vacuum between the molding and the plastic film) onto the molding (thermoforming). In general, an adhesive establishes a permanent bond between the plastic film and the molding.

However, a process such as this, which can be efficiently carried out with machine assistance, requires a thermoplastic film as the coating material. Hitherto, however, it has not been possible to finish leather-like substitute materials, for example LEFAs, in such a way that, on the one hand, they show the thermoplastic properties which are required for use in thermoforming processes but, on the other hand, still have leather-like surface properties.

WO 98/50617 relates to a thermoplastic composite material which contains, for example, leather and a binder, the binder containing polyacrylates of at least one aqueous anionic polyacrylate dispersion and at least one aqueous cationic polyacrylate dispersion. The described composite materials have the disadvantage of limited cohesion so that, for example where the described composite materials are used in thermoforming processes, only limited profile depths (radii) can be coated without the composite material tearing.

Accordingly, the problem addressed by the present invention was to provide a composite material which would largely exhibit the properties of the natural fibers used as the base material, but which on the other hand would be suitable for use in modern thermoforming processes by virtue of its thermoplastic properties, would have sufficient cohesion for the crack-free coating and "bridging" of large profile depths and which, in addition, would be easy to produce.

Another problem addressed by the present invention was to provide an adhesive with which the thermoplastic composite material could be firmly and durably bonded to a number of surfaces in the course of

machine processing.

It has now been found that a thermoplastic composite material containing organic fibers material, more particularly leather fibers, and a thermoplastic binder as the matrix material can be obtained providing the
5 matrix material used is a thermoplastic binder which contains at least 10% by weight of a polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl acetate copolymers or mixtures or copolymers of two or
10 more of the polymers mentioned, the matrix material not consisting solely of two polyacrylates.

The thermoplastic composite material obtainable from the polymers mentioned above preferably has a flow transition limit of about 70 to about 130°C.

15 Accordingly, the present invention relates to a thermoplastic composite material containing

20 a) at least 15% by weight of an organic fibrous material or of a mixture of two or more organic fibrous materials as component A

and

b) at least 15% by weight of a thermoplastic binder as component B,

25 the binder containing at least 10% by weight of a polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl acetate copolymers or mixtures or copolymers of two or more of the polymers mentioned, the binder not
30 consisting solely of two different polyacrylates.

Component A of the thermoplastic composite material may be any organic fibrous material which provides the thermoplastic composite material with the properties required by the user, for example a certain appearance or a certain feel. Organic fibrous materials in the context of 5 the present invention are understood to be both naturally obtained or naturally obtainable fibers and synthetic fibers providing they have an "organic base". Accordingly, organic fibrous materials do not include, for example, such fibers as asbestos, glass fibers or carbon fibers.

In addition, no distinction is made in the present specification 10 between materials which occur naturally in fibrous form and materials which have to be given a fibrous structure by a certain treatment step. In addition, among the natural materials, both vegetable and animal organic fibrous materials are suitable for the purposes of the invention.

Synthetic fibers, vegetable fibers or animal fibers are normally used 15 for the purposes of the invention.

Suitable natural fibers include, for example, animal fibers, such as wool, hair or silk. Other suitable natural fibers are vegetable fibers, for example cotton, kapok, flax, hemp, jute, kenaf, ramie, broom, abaca, coconut or sisal. Suitable synthetic fibers of natural polymers are cupro 20 fibers, viscose fibers, modal fibers, acetate fibers, triacetate fibers and protein fibers or alginate fibers or mixtures of two or more of the fibers mentioned.

Suitable fibers of synthetic polymers are, for example, polyacrylic, polymethacrylic, polyvinyl chloride, fluorine-containing polymer fibers, poly-25 ethylene, polypropylene, vinyl acetate, polyacrylonitrile, polyamide, polyester and polyurethane fibers.

However, it is particularly preferred to use leather fibers as the organic fibrous material. To obtain these fibers, leather waste is fiberized and size-reduced by a suitable process so that the fibers obtained can be 30 subsequently used in the process according to the invention for producing

a thermoplastic composite material with leather-like properties.

Basically, the leather fibers may be obtained from any type of leather waste. The leather waste may originate both from chrome-tanned leather and from vegetable-tanned leather. Leathers suitable for use in accordance with the invention include, for example, box leathers, such as hide box, calf box or mast box, buffed box, glazed kid, suede, kid, sandal leather, sole leather, lining leather, nappa leather, glove leather, buckskin, harness leather and industrial leather, chamois leather, hatband leather or transparent leather.

Depending on the decorative or mechanical effect required, the organic fibrous material may be size-reduced to a stretched length of about 0.1 to 15 mm. Where leather fibers in particular are used, fiber lengths of about 0.5 to 13 mm are suitable, fiber lengths of about 1 mm to about 10 mm being preferred and fiber lengths of about 3 to about 8 mm particularly preferred. The fiber length is measured in the stretched state of the fibers. Depending on the starting material used and the size reduction method applied, the fibers may of course assume an irregularly curved shape in the absence of any external influence.

Component A is present as the base material of the thermoplastic composite material according to the invention in a quantity of at least about 10% by weight. The thermoplastic composite material increasingly assumes the properties of the organic fibrous material with increasing percentage content of component A. Accordingly, depending on the desired effect, it can be of advantage to use, for example, at least 20% by weight or at least about 25% by weight of component A in the thermoplastic composite material according to the invention. However, the percentage content of the organic fibrous material may even be greater, for example about 30% by weight, 35% by weight, 40% by weight, 45% by weight or even more than about 50% by weight, percentage contents of for example 55% by weight or even 60% by weight and more being possible. In one

particularly preferred embodiment, the percentage content of fibrous materials is about 15 to about 50% by weight, percentage contents of about 25 to less than about 45% by weight being most particularly preferred.

5 Leather fibers are preferably present as component A in the thermoplastic composite material according to the invention.

In order to provide the thermoplastic composite material with the thermoplastic properties required for further processing, the thermoplastic composite material contains a thermoplastic binder as component B.

10 Binders in the context of the present invention are understood to be polymeric compounds which serve as the matrix of the composite material. Polymeric materials with a molecular weight of more than about 1,000 are generally used as binders. However, the molecular weight is preferably higher.

15 The molecular weight (M_n) of the polymers present in the binder is preferably in the range from about 10,000 to about 1,000,000, more preferably in the range from about 20,000 to about 300,000 and most preferably in the range from about 50,000 to about 150,000.

20 In the context of the present invention, the term "binder" is used for the polymeric matrix material as a whole, irrespective of the number of its constituent polymer components and irrespective of how many different preparations containing the polymers forming the binder were required for its production.

25 The molecular weight distribution of the polymers, which can be determined for example by gel permeation chromatography (GPC), does not have to be monomodal. If desired, the thermoplastic binder may also have a bimodal or higher modal distribution.

30 A binder containing at least 10% by weight of a polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin

copolymers, polyacrylates or ethylene/vinyl acetate copolymers or mixtures or copolymers of two or more of the polymers mentioned, the matrix material not consisting solely of two polyacrylates, is used for the production of the thermoplastic composite material according to the
5 invention.

In a preferred embodiment of the invention, binders containing at least two different polymers are used for the production of the thermoplastic composite materials. "Two different polymers" in the context of the invention are understood to be two polymer types which differ in their
10 chemical composition, i.e. in the nature of the monomers involved in the synthesis of the polymer or - where two or more monomers are involved in the synthesis of the polymer - in the ratio of those monomers to one another or in both. It does not matter whether the individual polymer has thermoplastic properties as long as the mixture of two different polymers
15 has corresponding thermoplasticity.

Polyurethanes in the context of the present invention are understood to be any polymers which contain at least two urethane groups in the polymer backbone. Polyurethanes suitable for the purposes of the present invention are any thermoplastic polyurethanes known to the expert on
20 polyurethane chemistry, more especially polyurethanes of the type normally used in the production of thermoplastic moldings, more particularly films, or for the thermoplastic coating of surfaces. Suitable polyurethanes are, for example, polyester polyurethanes or polyether polyurethanes obtainable by reaction of dicarboxylic acids with
25 corresponding polyhydric alcohols, more especially dihydric alcohols, for example dihydric polyethers, such as polyethylene oxide, to form polyether or polyester polyols and subsequent reaction of the corresponding polyether or polyester polyols with di- or polyfunctional isocyanates.

Polyesters in the context of the present invention are any polymers
30 containing at least two ester groups and no urethane groups in the polymer

backbone. Polyesters suitable for the purposes of the present invention are any thermoplastic polyesters known to the expert, more especially polyesters of the type normally used in the production of thermoplastic moldings, more particularly films, or for the thermoplastic coating of surfaces. Suitable polyesters are, for example, the polyesters obtainable by reaction of dicarboxylic acids with corresponding polyhydric alcohols, more especially dihydric alcohols, for example difunctional polyethers, such as polyethylene oxide.

Compounds suitable for the production of corresponding polyurethanes or polyesters are mentioned hereinafter.

Polyamides in the context of the present invention are any thermoplastic polyamides of the type obtainable by reaction of suitable di- or polycarboxylic acids with corresponding amines.

Polyolefins suitable for the purposes of the present invention are obtainable, for example, by radical or co-ordinative polymerization of α -olefins, more especially ethylene or propylene.

Polyvinyl esters suitable for the purposes of the present invention are, in particular, polymers of vinyl acetate.

Polyethers suitable for the purposes of the invention are, for example, polyethylene oxide, polypropylene oxide, polybutylene oxide or polytetrahydrofuran, more particularly with a molecular weight of more than about 5,000.

Suitable polystyrenes are, for example, the polymers of styrene or α -methyl styrene.

Other polymers suitable for use in the binder of the thermoplastic composite material according to the invention are the styrene/olefin copolymers obtainable by copolymerization of styrene with mono- or diolefins, more especially butadiene. A preferred embodiment of the invention is characterized by the use of the styrene/butadiene copolymers obtainable, for example, under the name of Kagetex LBS 3565 or Kagetex

LBS 3060 from Kautschukgesellschaft or under the name of Intex 131 from Enichem or under the name of 28 W 20 from Synthomerchemie.

Suitable polyvinyl esters are the polymers of the esters of unsaturated alcohols with corresponding carboxylic acids. Suitable unsaturated alcohols are, for example, unsaturated aliphatic alcohols containing 2 to about 22 carbon atoms, more especially 2 to about 8 carbon atoms. Suitable carboxylic acids are linear and branched alcanoic acids containing 2 to about 22 carbon atoms, more especially 2 to about 8 carbon atoms. A preferred embodiment of the invention is characterized by the use of polyvinyl acetate. Suitable polyvinyl acetates are obtainable, for example, under the name of Vinnapas D50 from Wacker, Mowilith D 60 from Clariant or Vinnamul 60 044, Vinnamul 9300, Vinnamul 9960 or Vinnamul 84125 from Vinnamul (NL).

Polymers present in the form of an aqueous dispersion are preferably used for the production of the composite materials according to the invention. The dispersions may be anionically stabilized or cationically stabilized polymer dispersions. The dispersion may be stabilized, for example, by self-emulsifiable polymers, i.e. by polymers bearing corresponding hydrophilic groups, for example carboxylic acid groups or amino groups. However, dispersions stabilized by suitable anionic or cationic dispersants or emulsifiers may also be used.

The terms "polyacrylate" and "polyacrylates" used in the present text apply in the following both to polymers or copolymers of acrylic acid and/or derivatives thereof and to polymers or copolymers of methacrylic acid and/or derivatives thereof.

Polyacrylates can be produced by subjecting acrylic acid and/or methacrylic acid and/or derivatives of acrylic acid and/or methacrylic acid, for example esters thereof with monohydric or polyhydric alcohols, either on their own or in the form of mixtures of two or more thereof in known manner to polymerization, for example radical or ionic polymerization.

Polyacrylates in the form of an anionic dispersion obtainable, for example, by emulsion polymerization of the corresponding monomers and comonomers are preferred for the purposes of the invention. In general, aqueous anionic dispersions contain, for example, the sodium, potassium and/or ammonium salts of long-chain aliphatic carboxylic acids and/or sulfonic acids for emulsification. However, alkali metal C₁₀₋₁₈ alkyl sulfates, ethoxylated and sulfated and/or sulfonated long-chain aliphatic alcohols or alkylphenols and sulfodicarboxylic acid esters are also suitable.

Homopolymers or copolymers which, besides the acrylates, also contain styrene, acrylonitrile, vinyl acetate, vinyl propionate, vinyl chloride, vinylidene chloride and/or butadiene may be used as polyacrylates in accordance with the present invention.

Monomers suitable for use in the production of the polyacrylates are, in particular, methyl acrylate, ethyl acrylate, n-butyl acrylate, isobutyl acrylate, tert.butyl acrylate, hexyl acrylate, 2-ethylhexyl acrylate and lauryl acrylate. Acrylic acid, methacrylic acid, acrylamide or methacrylamide may optionally be added in small quantities as further monomers during the polymerization reaction.

Other acrylates and/or methacrylates containing one or more functional groups may optionally be present during the polymerization reaction. Examples of these other acrylates and/or methacrylates are maleic acid, itaconic acid, butanediol diacrylate, hexanediol diacrylate, triethylene glycol diacrylate, tetraethylene glycol diacrylate, neopentyl glycol diacrylate, trimethylol propane triacrylate, 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate, hydroxypropyl acrylate, propylene glycol methacrylate, butanediol monoacrylate, ethyl diglycol acrylate and, for example, 2-acrylamido-2-methyl propane sulfonic acid as a monomer containing sulfonic acid groups.

Acrylate/vinyl ester copolymers, acrylate/styrene copolymers and acrylate/methacrylate copolymers are particularly preferred. Polyacrylate

dispersions of the type marketed by BASF AG, Ludwigshafen, under the registered name of Acronal®, more particularly the products Acronal® 500 D and Acronal® S312 D, are particularly preferred and suitable for use in the thermoplastic composite materials according to the invention.

5 In general, anionic polyacrylate dispersions contain only one type of polymer. If two different polyacrylates from anionic dispersions are to be used, it will generally be necessary to use two different anionic dispersions which contain the particular polymer in dispersed form. According to the invention, however, it is immaterial whether the different polyacrylates
10 preferably present in the matrix are present in a single dispersion or whether they have been obtained from two or more different dispersions.

In a preferred embodiment of the invention, the binder used as component B contains at least two of the above-mentioned polymers, the total percentage content of the two polymers in the binder as a whole, i.e.
15 in component B as a whole, being at least about 10% by weight, for example at least about 20% by weight, at least about 30% by weight or at least about 40% by weight or more, for example at least about 50 to about 70% by weight or up to about 80% by weight.

20 In another preferred embodiment of the invention, the binder used as component B contains only one polyacrylate besides two of the above-mentioned polymer types or no polyacrylate.

In another preferred embodiment of the invention, the binder contains at least one polyvinyl acetate and at least one styrene/butadiene copolymer, more particularly a corresponding block copolymer.

25 It has also been found that the thermoplastic properties of the thermoplastic composite material according to the invention can be influenced by choosing polyacrylates having a suitable minimum film forming temperature. The minimum film forming temperature of a polymer is the lowest temperature at which a dispersion still just forms a coherent
30 film after evaporation of the water. It is close to the glass transition

temperature T_g of the polymer and with film formation determines one of the most important performance properties of a polymer dispersion. The minimum film forming temperature (MFT) is generally determined in accordance with DIN 53787. The measuring instrument used is a metal plate to which a temperature gradient is applied. The film is observed for the temperature at which it begins to develop cracks or for the location of the so-called white point at which the cloudy film begins to become clear.

In a preferred embodiment of the invention, at least one of the polymers has an MFT of up to 30°C.

10 In another preferred embodiment of the invention, at least one of the polymers has an MFT of about up to 5°C and at least one other polymer has an MFT of about 15°C to about 25°C.

15 Component B as a whole preferably makes up at least about 20% by weight of the thermoplastic composite material. It can be of advantage, for example for selective property modifications, if the thermoplastic composite material contains at least about 30% by weight or at least about 40% by weight or more of component B, for example at least about 50 to at least about 80% by weight. In a preferred embodiment of the invention, component B makes up about 50 to about 75% by weight of the thermoplastic composite material as a whole.

20 Besides the organic fibrous material as component A and the thermoplastic binder as component B, the thermoplastic composite material according to the invention may contain other components, preferably in a quantity of up to about 20% by weight. These other components include, for example, inorganic salts, cationic polymers, preservatives, dyes, natural and/or synthetic fats, paraffins, natural and/or synthetic oils, silicone oils and ionic and/or nonionic surfactants.

25 Salts of aluminium or copper are preferably used as the inorganic salts, aluminium sulfate being particularly preferred.

30 The inorganic salts are generally used in the production process,

which will be described hereinafter, to precipitate (coagulate) the polymeric binder. In general, most of the metal salt is removed from the composite material with the aqueous phase although a small residue can remain in the composite material.

- 5 Cationic polymers or cationically charged polymers are polymers which carry positive charges on the polymer backbone or on side groups covalently bonded to the polymer backbone.

- Cationic charges can be applied to a polymer by application of suitable cationic groups. "Cationic groups" are understood to be groups
10 which either carry a positive charge or which can be converted into a group carrying a cationic charge by a simple reaction, for example quaternization. In general, the groups in question are amine or ammonium groups.

- "Quaternization" is understood to be a reaction in which a primary, secondary or tertiary amine is converted into the tetravalent positively
15 charged form by reaction with a suitable reagent. Suitable reagents are, for example, alkyl halides, more especially alkyl chlorides, alkyl bromides and alkyl iodides, and protonic acids, for example mineral acids, such as hydrohalic acid, phosphoric acid, sulfuric acid, or strong organic acids, such as formic or acetic acid.

- 20 Suitably modified polyacrylates, polyesters, polyurethanes or other polymers carrying at least one primary, secondary or tertiary nitrogen atom may be used as the polymers bearing cationic groups. Polyacrylates bearing cationic groups are preferably used for the purposes of the present invention.

- 25 Corresponding polyacrylates can be produced, for example, by copolymerizing the monomers described above (except for the monomers bearing free acid groups) together with a suitable ethylenically unsaturated compound bearing a primary, secondary or tertiary amino group. Suitable ethylenically unsaturated monomers containing primary, secondary or
30 tertiary amino groups are, for example, dimethylaminoethyl methacrylate,

dimethylaminoneopentyl acrylate, dimethylaminopropyl methacrylate, aminoethyl methacrylate, 2-N-morpholinoethyl acrylate, tert.butylaminoethyl methyl acrylate, 4-methacrylamido-2,2,6,6-tetramethyl piperidine, trimethyl ammonium methyl (meth)acrylate chloride, α -acetamidodiethyl aminoethyl (meth)acrylate chloride, trimethyl ammonium propyl acrylate chloride or methacrylate chloride, trimethyl ammonium methyl acrylate bromide or methacrylate bromide, trimethyl ammonium neopentyl methacrylate chloride or acrylate chloride, diallyl dimethyl ammonium chloride, diallyl butyl methyl ammonium bromide. The cationically charged polymers 5 preferably have a pH value of about 3 to about 8 although, in one particularly preferred embodiment, the pH value of the cationically charged polymers is neutral, i.e. in the range from about 6.5 to about 7.5.

The charge density, i.e. the number of cationic groups per unit weight in the polymer, is preferably about 2 to about 6 meq/g and, in one 15 particularly preferred embodiment, is in the range from about 2.5 to about 5 meq/g. Most particularly preferred charge densities are in the range from about 3.5 to about 4.5 meq/g.

The cationic polymer generally has a molecular weight of about 1,000 to about 500,000, molecular weights of about 50,000 to about 20 150,000 being preferred and those in the range from about 80,000 to about 120,000 being particularly preferred.

Preferred preservatives are those which have a fungicidal action spectrum. The preservative Preventol® A11D marketed by Bayer AG, Leverkusen, is particularly suitable for the purposes of the present 25 invention.

In one particularly preferred embodiment, component B contains at least 50% by weight of polyvinyl acetate.

In another preferred embodiment of the invention, component B contains at least 20% by weight of a copolymer of butadiene and styrene.

30 In another preferred embodiment, the thermoplastic composite

material contains

- about 10 to about 50% by weight of organic fibers,
- about 10 to about 40% by weight of at least one copolymer of styrene and butadiene,
- about 30 to about 60% by weight of polyvinyl acetate and
- optionally up to 25% by weight of inorganic salts, preservatives, dyes, natural and/or synthetic fats, paraffins, natural and/or synthetic oils, silicone oils, ionic and/or nonionic surfactants.

10

The thermoplastic composite material according to the invention is preferably intended for coating surfaces having a relief structure. Such coatings are normally applied by so-called thermoforming. In a preferred embodiment of the invention, the thermoplastic composite material has a flow transition range of about 70 to 130°C.

At temperatures above the flow transition range mentioned, the thermoplastic composite materials according to the invention can undergo irreversible dimensional changes, for example elongation. By virtue of its particular tear strength coupled with its good flow behavior, the thermoplastic composite material according to the invention is even able to coat reliefs with considerable differences in height (radii) without cracks in the coating and without any change in the original surface structure.

The thermoplastic composite material according to the invention is preferably produced by contacting component A with the constituents of component B, these constituents preferably being present in the form of an aqueous dispersion.

If component B contains more than one constituent, i.e. more than one polymer, both polymers may be present alongside one another in one and the same dispersion. However, in another embodiment of the invention, the two polymers may be present in different dispersions.

In the production process according to the invention, component A and the constituents of component B are mixed in one or more dispersions and the constituents of component B are coagulated simultaneously, i.e. during mixing, or subsequently, i.e. in a separate process step after mixing.

5 Accordingly, the present invention also relates to a process for the production of a thermoplastic composite material containing

- 10 a) at least 15% by weight of an organic fibrous material or of a mixture of two or more organic fibrous materials as component A
and
b) at least 15% by weight of a thermoplastic binder as component
B,

in which fibers with a stretched fiber length of 0.1 to 15 mm as component
15 A are mixed simultaneously or successively in any order with a polymer dispersion or a mixture of two or more polymer dispersions each containing at least one polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl
20 acetate copolymers or mixtures or copolymers of two or more of the polymers mentioned, the dispersion(s) not consisting solely of two different polyacrylates, to form a mixture so that the polymers present in the dispersion(s) form component B, after which the mixture is treated with an aqueous solution of an aluminium salt or a copper salt, dewatered and
25 dried.

If two or more different polymer dispersions are used, differently stabilized dispersions, for example, may be used in accordance with the invention. For example, where two polymer dispersions are used, an anionically stabilized dispersion and a cationically stabilized dispersion may
30 be used. The dispersions may be selected so that substantially complete

coagulation, i.e. substantially complete precipitation of the binders present in the dispersion, occurs. However, the binders may also be only partly precipitated.

However, a preferred embodiment of the invention is characterized
5 by the use of polymer dispersions which are stabilized substantially identically, at least in regard to the charge of the stabilizing species. For example, anionically or cationically stabilized dispersions may be used. In another preferred embodiment of the invention, anionically stabilized polymer dispersions are used.

10 The treatment of the mixture with an aqueous solution of an aluminium or copper salt is conducted in such a way that, after the treatment, substantially all polymer molecules present in the mixture are precipitated, i.e. coagulated.

15 The following exemplary description of the process is merely intended to illustrate one way of carrying out the process according to the invention and does not represent a limitation. Modifications of subsequent process steps, optionally with a view to optimizing the process described hereinafter, may readily be undertaken by the expert according to the situation confronting him.

20 To produce the thermoplastic composite materials according to the invention, tanned leather waste is size-reduced (precut) in cutter mills to a size of about 1 cm². At this stage, size reduction is generally carried out under dry conditions.

25 The leather waste thus precut is weighed and then wet-fiberized in so-called disk refiners. The addition of water is controlled in such a way that a lump-free fiber slurry consisting of about 5% by weight of fibers and about 95% by weight of water is formed (corresponding to about 1,000 kg of fibers to 20 m³ of water). The size reduction step is preferably carried out in such a way that part of the wastewater subsequently formed in the
30 process is returned to the circuit at this point. In this way, around 50% and

preferably more of the water required for the size reduction step is made up of wastewater.

The suspension of leather fibers in water obtainable in this way is then transferred to a suitable vessel, preferably a mixing vat. The quantity 5 transferred is gauged in such a way that the concentration of leather fibers, based on the mixture as a whole, is between about 1.5 and about 2.5% by weight.

If the leather fibers contain chrome-tanned leather or if the leather fibers consist solely of chrome-tanned leather, vegetable tanning agents, 10 for example chestnut wood extract, quebracho, mimosa or valonea, are first added.

Oiling agents are then added. Suitable oiling agents are any leather oiling agents which are emulsifiable in water. Sulfated fish oils are preferably used, for example Licrol® DM 10, a product of Erberle, Coriatol 15 U6, a product of Polychemie, or Coripol 2397, a product of TFL.

Dyes may also be added to the mixture. The dyes used are normally quantitatively absorbed iron oxide dyes, for example Bayferrox® 960 (a product of Bayer AG).

Preservatives, for example Preventol A11D (Bayer AG), natural 20 and/or synthetic fats, natural and/or synthetic oils, silicone oils and/or ionic and/or nonionic surfactants, may also be added to the mixture.

If the additives are insoluble in water or do not self-emulsify in water, they are generally added to the mixture in the form of emulsions.

The polymer dispersion or the mixture of two or more different 25 polymer dispersions is then added.

If two or more different polymer dispersions are to be used, they may be added to the mixture either simultaneously or successively in any order.

If two polymer dispersions that are differently stabilized, i.e. with 30 different charges of the stabilizing species, are added to the mixture, the

anionically stabilized polymer dispersion(s) is/are added separately from the cationically stabilized polymer dispersion(s). The order in which the dispersions are added is of no consequence.

After the polymer dispersions have been added, a solution of an aluminium salt or a copper salt is added to the mixture. Aluminium sulfate is preferably used. About 40 to 300 l, preferably about 100 to about 250 l and, in a particularly preferred embodiment, about 120 to about 200 l of an approximately 20 to preferably about 60% by weight aluminium sulfate solution are added per 1,000 kg of mixture.

During but preferably after the metal salt treatment, the mixture may be treated with a solution of a cationically charged polymer. The cationically charged polymer preferably has a molecular weight of no more than about 150,000 and advantageously in the range from about 80,000 to about 120,000.

The charge density of the cationically charged polymer is about 2.00 to 5.00 meq/g, preferably in the range from about 2.50 to about 4.50 meq/g and more preferably in the range from about 3.00 to about 4.00 meq/g.

The cationically charged polymer is used in a quantity of about 0.1 to 10% by weight, preferably in a quantity of about 1 to 8% by weight and more preferably in a quantity of about 2 to 6% by weight, based on the total solids in the mixture.

After stirring for about 1 hour, the mixture is freed from excess water using a suitable drainage machine. Although drainage machines operating on the so-called batch principle, for example a so-called Müller press, may be used for this purpose, continuous drainage in a fourdrinier machine is preferred. The mixture is drained to a residual water content of around 70% by weight in the fourdrinier machine.

After the drainage step, the material obtained is pressed in a suitable press, preferably an intermittent press, under a pressure of about 1,000 to about 3,000 t and preferably in the range from about 1,500 to

about 2,500 t.

The material thus treated is then passed through a suitable thermal dryer, in which it is dried to a residual water content of about 10% by weight, and thereafter is wound onto rolls.

5 The films obtained in this way have a flow transition range of from about 70 to about 130°C and are thus suitable for modern surface coating processes, preferably so-called thermoforming processes.

An adhesive is generally used to establish a permanent bond between the surface coating material and the surface to the coated.

10 Adhesives in the context of the present invention are understood to be non-metallic, preferably organic materials which bond parts to be joined by surface adhesion and internal strength.

Suitable adhesives include, for example, glues, dispersion adhesives, solvent-based adhesives and/or contact adhesives.

15 Organic adhesives - either physically setting adhesives or chemically reacting adhesives or a combination of the two - are preferably used for the purposes of the invention. Physically setting adhesives which may be used for the purposes of the invention include, example, dissolved or dispersed adhesives, contact adhesives or hotmelt adhesives. Suitable chemically reacting adhesives are, for example, those which impart adequate adhesion to the surfaces to be joined to one another without emitting volatile constituents. However, it is also possible to use adhesives which produce the necessary adhesion while emitting volatile constituents. The adhesives may be both cold-curing and hot-curing adhesives, may have a 20 thermoplastic, thermoset or elastomeric end state and may be applied as 25 one-component adhesives or as two- or multi-component adhesives.

In general, it is of advantage to expose the surfaces to be joined to a high pressure after they have been fitted together. Accordingly, one particular embodiment of the present invention is characterized by the use 30 of adhesives which, after application of pressure to the surfaces to be

joined together, produce improved adhesion in relation to pressureless processing.

The adhesives used in accordance with the present invention advantageously contain at least three different macromolecular compounds
5 as coupling agents.

Coupling agents in the context of the present invention are macromolecular compounds which mainly contribute towards the generation of the adhesion and cohesion forces between the surfaces to be bonded.

10 According to the invention, a combination of polyurethane, polyacrylate and ethylene/vinyl acetate copolymer (EVA copolymer), for example, in the form of an aqueous dispersion or hotmelt adhesive is used as the coupling agent.

15 More particularly, the adhesive according to the invention contains about 30 to 120 parts by weight of a preferably anionic polyester polyurethane dispersion.

Polyester polyurethanes can be obtained by reacting generally relatively low molecular weight OH-terminated polyesters with at least difunctional isocyanates to form urethane links. The polyesters generally
20 have a molecular weight of at least about 400 and preferably of at least about 500. Both aromatic and aliphatic polyesters may be used although aromatic polyesters are normally not suitable as the sole polyester component in view of their marked tendency to crystallize and their lack of flexibility.

25 In general, the acid components involved in the synthesis of the polyester are, for example, aliphatic dicarboxylic acids, such as butane-1,4-dicarboxylic acid, pentane-1,5-dicarboxylic acid, hexane-1,6-dicarboxylic acid, heptane-1,7-dicarboxylic acid and other higher homologs with an increasing number of carbon atoms. In addition, aromatic dicarboxylic
30 acids, for example terephthalic acid or isophthalic acid, may be used.

In general, any aliphatic dialcohols containing 2 to about 20 carbon atoms may be used as the diol component for the production of the polyesters. Preferred dialcohols are ethylene glycol, propylene glycol, butylene glycol, pentane-1,5-diol, hexane-1,6-diol, heptane-1,7-diol, 5 octane-1,8-diol, nonane-1,9-diol, decane-1,10-diol and higher homologs thereof containing up to 20 carbon atoms. Aliphatic diols containing 2 to 10 carbon atoms and preferably about 4 to 8 carbon atoms are preferably used.

The polyesters may also have been produced using acid or alcohol components with a functionality different from 2. Suitable acid components with a functionality of 3 or more are, for example, 1,2,3-propane tricarboxylic acid, hemimellitic acid, trimellitic acid, trimesic acid and 1,2,4,5-benzenetetracarboxylic acid. Trihydric or higher alcohol components are, for example, glycerol, trimethylol propane, pentaerythritol 15 and carbohydrates such as, for example, the monomeric sugar compounds, more especially glucose.

The reaction products of dihydric or higher alcohols with ethylene oxide and/or propylene oxide may also be used as the diol component.

Basically, mixtures of several different acids and several different alcohols may also be reacted with one another.

The reaction generally proceeds with a stoichiometry which, on the one hand, enables the required molecular weight to be established (generally about 400 to about 10,000 and preferably about 500 to about 5,000) and, at the same time, ensures that the polyester contains at least 25 one hydroxy group in the molecule, preferably as the terminal group.

The described polyesters are then reacted with isocyanates having a functionality of two or more, for example with triisocyanates or tetra-isocyanates. In principle, any aliphatic or aromatic polyisocyanates may be used for this purpose. Preferred polyisocyanates are, for example, 30 hexamethylene-1,6-diisocyanate, 2,4- and 2,6-toluene diisocyanate and

4,4'-diisocyanatodiphenyl methane (MDI). If toluene diisocyanate is used, it is preferably used in the form of an isomer mixture in which the 2,4- and 2,6-isomers are present in a ratio of about 80 to about 20.

The polyurethanes are preferably present in dispersed form, water 5 preferably being the continuous phase. Anionic emulsifiers are generally used.

In one particularly preferred embodiment, the polyurethane dispersions have a solids content of about 35 to about 65% and a viscosity (according to DIN 53019) of less than about 1,000 mPas, preferably in the 10 range from about 100 to about 900 mPas and more preferably in the range from about 200 to about 800 mPas. The pH value is preferably in the range from about 5 to about 9 and more preferably in the range from about 6.5 to about 7.5. The minimum film forming temperature (DIN 53787) of the preferred polyurethane dispersions is between about 2 and about 10°C, 15 preferably between about 4 and about 6°C and, in a particularly preferred embodiment, around 5°C. The softening point of the films obtainable from these polyurethane dispersions may be above 100°C, as determined in accordance with ASTM D 816, but is preferably of the order of 80°C or lower, for example around 60°C.

20 Examples of commercially available polyurethane dispersions suitable for use in accordance with the invention are Dispercoll® U 42, Dispercoll® U 53, Dispercoll® U 54, Dispercoll® KA 8481 (products of BAYER AG), Quilastic® DEP-172 and/or Quilastic® 144-66. Dispercoll® U 53 is particularly preferred.

25 Polyolefins or polyamides are also suitable adhesives and are used in particular as hotmelt adhesives.

The polyacrylates are also preferably used in the form of an aqueous dispersion. In one particularly preferred embodiment, the dispersion is based on anionic emulsifiers. The polyacrylate dispersions 30 are preferably polymer dispersions which, basically, may already be used

as adhesives, so-called acrylate adhesives. Polyacrylates are preferably based on acrylic acid esters, preferably methyl, ethyl, n-butyl, isobutyl or 2-ethylhexyl esters of acrylic acid. The corresponding esters of methacrylic acid may also be used together with or instead of the acrylic acid esters.

- 5 The acrylates and/or methacrylates may be copolymerized with other substances which contain an olefinically unsaturated double bond. In a preferred embodiment, these other substances are, for example, vinyl esters of carboxylic acids, for example vinyl acetate, or styrene.

The polyacrylates may optionally contain functional groups which 10 allow subsequent crosslinking, for example by an increase in temperature. The functional groups in question are preferably functional groups which can be crosslinked by adding a small quantity of a catalytically active substance and increasing the temperature. In a particularly preferred embodiment, an acid or any other compound which is capable of releasing 15 an acid by an increase in temperature in a suitable environment, for example an aqueous environment, is used as the catalyst. Such compounds as oxalic acid, ammonium chloride, magnesium chloride and/or diammonium phosphate are preferably used.

The polyacrylate dispersions preferably have a viscosity at 23°C 20 (DIN 53019) of about 20 to about 5,000 mPas. The pH value of the preferred dispersions is generally in the range from about 2 to about 9 and preferably in the range from about 2.5 to about 8. All the polyacrylate dispersions used have a minimum film forming temperature (DIN 53787) below about 1°C and preferably below about 0°C. The polyacrylates 25 generally have a glass transition temperature of less than about -20°C, preferably less than -30°C and, in a particularly preferred embodiment, less than -35°C, for example in the range from -40°C to -45°C. The average particle size of the dispersed polyacrylate particles is preferably less than about 70 µm and, more preferably, less than about 65 µm.

30 Preferred commercial products are obtainable, for example, from

BASF under the name of Acronal® V 205, from Merquinsa under the Quiacryl® 126-07 or from Röhm under the name of Plextol® E 220.

EVA copolymers are copolymers of ethylene and vinyl acetate. The two monomers can be copolymerized in any quantity ratios using radical initiators. The copolymers obtained are characterized by a statistical distribution of the monomer units in the polymer chains. The properties of the EVA copolymers may be varied within wide limits through the molar ratio of ethylene to vinyl acetate. For example, products with an ethylene content of less than 30% by weight are partly crystalline and thermoplastic while products with a vinyl acetate content of about 40 to about 70% by weight are substantially amorphous. The EVA copolymers are generally produced by bulk, emulsion or solution polymerization. The molecular weight of the EVA copolymers used in accordance with the invention is in the range from about 10,000 to about 1,500,000. The EVA copolymers are preferably used in the form of an aqueous dispersion with a solids content of about 40 to about 70% by weight and preferably in the range from about 50 to about 60% by weight. The dispersion has a viscosity (to ISO 2555 at 23°C, Brookfield (RVT), 20 r.p.m., spindle 4) in the range from about 2,000 to about 13,000 mPas. For example, dispersions with a viscosity in the range from about 4,000 to about 6,000 mPas, from about 6,000 to about 12,000 mPas or from about 2,500 to about 4,000 mPas may be used in accordance with the invention. The dispersions have a pH value of about 3 to about 6 and preferably in the range from about 3.8 to about 4.8. The dispersions produce opaque, transparent or clear films and generally have a minimum film forming temperature of less than 5°C, preferably less than about 3°C and, in a particularly preferred embodiment, around 0°C. The films have a tear strength (standard conditioning atmosphere of 23°C/50% relative humidity to DIN 50014) of about 2.5 to about 9 N/mm². For example, dispersions which produce films having a tear strength of about 3 N/mm², 6 N/mm² or about 8 N/mm² may be used in accordance with the

invention. The films produced from the dispersions should have an elongation at break (as determined to DIN 50014 in a standard conditioning atmosphere of 23°C/50% relative humidity) of about 500 to about 900%. The films should have a cold crack temperature (DIN 53372) of at most 5 about 4°C.

The adhesive according to the invention may contain other components, for example wetting agents, thickeners, preservatives or defoamers.

Wetting agents are used to improve substrate wetting in aqueous systems. In the present case, wetting agents improve the wetting of the thermoplastic composite material and of the surface to be bonded thereto. Basically, any surfactants which do not adversely affect the stability of the adhesive dispersion may be used as wetting agents, although polyether-modified dimethyl polysiloxanes are preferably used. The commercially available product BYK® 346, a product of BYK Chemie GmbH, or Lumiten® IRA is particularly suitable.

Organic high molecular weight substances which absorb water and swell in the process, finally changing into viscous solutions, are used as thickeners. Suitable thickeners are, for example, natural thickeners, such as agar agar, carrageen, tragacanth, gum arabic, alginates, pectins, polyoses, guar gum, locust bean gum, starch, dextrans, gelatine or casein. Modified natural materials such as, for example, carboxymethyl cellulose and other cellulose ethers, hydroxyethyl and hydroxypropyl cellulose and gum ethers may also be used.

25 Organic synthetic thickeners, such as polycarboxylic acids, poly-
ethers, polyimines, polyamides and certain vinyl polymers, are also suitable
and are preferably used for the purposes of the present invention. Such
thickeners as polyvinyl alcohol or polycarboxylic acids, for example
carboxyfunctional acrylate copolymers, or vinyl pyrrolidone copolymers are
30 preferred. The thickeners are used in the form of an aqueous solution or

aqueous dispersion with a solids content of generally about 20 to about 40% by weight and preferably in the range from about 25 to about 30% by weight. Commercially available thickeners which may be used for the purposes of the invention are, for example, Collacral® VL (BASF) or

5 Latekoll® D (BASF).

A suitable defoamer is, for example, Agitan® 281 (Münzing Chemie).

A suitable preservative is, for example, Aktizid® RS (Thorchemie).

In another preferred embodiment of the invention, the adhesives
10 mentioned above are used as suitably formulated hotmelt adhesives.

The adhesive is applied to the substrate, preferably a presspahn board, to be coated with the thermoplastic composite material by means of a spray gun or any other suitable tool.

After a drying time of not less than 30 minutes, the sections of the
15 thermoplastic composite material according to the invention are placed onto the substrate to be coated and pressed at a temperature of about 80 to about 100°C. The pressing time is between about 10 and about 240 seconds and preferably between about 30 and about 120 seconds, depending on the thickness of the section. The pressure applied during
20 pressing is in the range from about 2 to about 10 bar and preferably in the range from about 3 to about 6 bar.

If the above-mentioned adhesives are used as hotmelt adhesives, the drying time mentioned is no longer necessary. Depending on the open time of the hotmelt adhesives, the sections should be contacted with the
25 substrate and pressed as quickly as possible. The use of hotmelt adhesives is particularly appropriate when extruded materials, for example extruded profiles, are to be coated continuously or discontinuously with the thermoplastic composite material according to the invention.

The present invention also relates to a process for the surface
30 coating of objects with the thermoplastic composite material according to

the invention in which the composite material according to the invention and the object are joined by hotmelt adhesive.

The present invention also relates to the use of a thermoplastic composite material according to the invention or a thermoplastic composite material produced in accordance with the invention for the surface coating of objects, more particularly for the profile sheathing of wall, floor and ceiling panels, furniture fronts with or without inner radii, for edge banding or for the surface coating of interiors of motor vehicles, for example dashboards, door mirrors, pillars, garnish moldings, gear levers, parcel shelves, trunk applications and the like.

The thermoplastic composite material is preferably applied by thermoforming in a vacuum. The thermoplastic composite material is preferably present in the form of films with a thickness of about 0.5 mm to about 1.0 mm and preferably with a thickness of about 0.6 mm to about 0.8 mm.

In another preferred embodiment of the invention, the thermoplastic composite material according to the invention has a surface coating which may be used, for example, to seal the material. Suitable surface coatings are, for example, thin films of polyurethane, polyester, polyolefins or the like which are laminated onto the surface of the thermoplastic composite material according to the invention using a suitable adhesive.

Accordingly, the present invention also relates to a thermoplastic composite material according to the invention which is laminated on at least one surface with a polymer film, more particularly of polyurethane, polyester or polyolefins.

In another preferred embodiment of the invention, the film contains dyes or pigments. In another embodiment of the invention, the film has a surface profile, for example in the form of pits or knobs.

CLAIMS

1. A thermoplastic composite material containing

- 5 a) at least 15% by weight of an organic fibrous material or of a mixture of two or more organic fibrous materials as component A
and
b) at least 15% by weight of a thermoplastic binder as component B,

10

characterized in that the binder contains at least 10% by weight of a polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl acetate copolymers or mixtures or copolymers of two or more of the polymers mentioned and does not consist solely of two different polyacrylates.

- 15 2. A composite material as claimed in claim 1, characterized in that it contains 15 to 45% by weight of organic fibrous material as component A.
3. A composite material as claimed in claim 1 or 2, characterized in that it contains at least 40% by weight of thermoplastic binder as component B.

- 20 4. A composite material as claimed in any of the preceding claims, characterized in that the thermoplastic binder contains at least 30% by weight of polyvinyl acetate.
25 5. A composite material as claimed in any of the preceding claims, characterized in that the thermoplastic binder contains at least 10% by weight of a copolymer of butadiene and styrene.
30 6. A composite material as claimed in any of the preceding claims, characterized in that it optionally contains up to 20% by weight of inorganic salts, cationic polymers, preservatives, dyes, natural and/or synthetic fats,

paraffins, natural and/or synthetic oils, silicone oils, ionic and/or nonionic surfactants.

7. A composite material as claimed in any of the preceding claims, characterized in that it contains synthetic fibers, vegetable fibers or animal fibers as component A.

8. A composite material as claimed in any of the preceding claims, characterized in that it contains leather fibers as component A.

9. A composite material as claimed in any of the preceding claims, characterized in that the fibers of component A have a stretched length of about 0.1 to about 15 mm.

10. A process for the production of a thermoplastic composite material containing

a) at least 15% by weight of an organic fibrous material or of a mixture of two or more organic fibrous materials as component A

and

b) at least 15% by weight of a thermoplastic binder as component B,

20 in which fibers with a stretched fiber length of 0.1 to 15 mm as component A are mixed simultaneously or successively in any order with a polymer dispersion or a mixture of two or more polymer dispersions each containing at least one polymer selected from the group consisting of polyurethanes, polyesters, polyamides, polyolefins, polyvinyl esters, polyethers, 25 polystyrenes, styrene/olefin copolymers, polyacrylates or ethylene/vinyl acetate copolymers or mixtures or copolymers of two or more of the polymers mentioned, the dispersion(s) not consisting solely of two different polyacrylates, to form a mixture so that the polymers present in the dispersion(s) form component B, after which the mixture is treated with an aqueous solution of an aluminium salt or a copper salt, dewatered and

dried.

11. The use of the thermoplastic composite material claimed in any of claims 1 to 9 or the thermoplastic composite material produced by the process claimed in claim 10 for the profile sheathing of wall, floor and ceiling panels, for the surface coating of furniture fronts with or without inner radii, for edge banding or for the surface coating of parts in interiors of motor vehicles.
5
12. A thermoplastic composite material as claimed in any of claims 1 to 9 or produced by the process claimed in claim 10, characterized in that it is laminated with a polymer film on at least one surface.
10
13. A process for the surface coating of objects with the thermoplastic composite material claimed in any of claims 1 to 9 or 12 or produced by the process claimed in claim 10, characterized in that the thermoplastic composite material and the surface of the object are bonded by hotmelt adhesive.
15

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INTERNATIONALE ANMELDUNG VERÖFFENTLICHT NACH DEM VERTRAG ÜBER DIE
INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS (PCT)

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(21) Internationales Aktenzeichen: PCT/EP00/03852		
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(71) Anmelder (<i>für alle Bestimmungsstaaten ausser US</i>): DORUS KLEBETECHNIK GMBH + CO. KG [DE/DE]; Kirchheimer Strasse 7-9, D-73441 Bopfingen (DE).	Veröffentlicht <i>Ohne internationalen Recherchenbericht und erneut zu veröffentlichen nach Erhalt des Berichts.</i>	
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(74) Anwalt: MATHES, Nikolaus; Henkel Kommanditgesellschaft auf Aktien, Patente (VTP), D-40191 Düsseldorf (DE).		

(54) Title: THERMOPLASTIC COMPOSITE MATERIAL

(54) Bezeichnung: THERMOPLASTISCHES VERBUNDMATERIAL

(57) Abstract

The invention relates to a thermoplastic composite material containing an organic fiber material and a mixture of two or more organic fiber materials and a thermoplastic bonding agent. The invention further relates to a method for producing said thermoplastic composite material and to the use of said thermoplastic material in the surface coating of objects.

(57) Zusammenfassung

Die Erfindung betrifft ein thermoplastisches Verbundmaterial enthaltend ein organisches Fasermaterial und ein Gemisch aus zwei oder mehr organischen Fasermaterialien und ein thermoplastisches Bindemittel. Weiterhin betrifft die Erfindung ein Verfahren zur Herstellung eines solchen thermoplastischen Verbundmaterials und die Verwendung des thermoplastischen Verbundmaterials zur Oberflächenbeschichtung von Gegenständen.

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PTO/SB/01 (6-95)

Approved for use through: 10/31/98 OMB 0651-0032

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Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

0010/PTO Rev. 6/95	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	H 4136 PCT/US
DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION		First Named Inventor	LORENZ, Juergen
COMPLETE IF KNOWN			
		Application Number	
		Filing Date	
		Group Art Unit	
		Examiner Name	

Declaration Submitted OR Declaration Submitted after Initial Filing

As a below named Inventor, I hereby declare that:
My residence, post office address, and citizenship are as stated below next to my name.
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

THERMOPLASTIC COMPOSITE MATERIAL

(Title of the Invention)

the specification of which

is attached hereto

OR

was filed on (MM/DD/YYYY) as United States Application Number or PCT International

Application Number and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	Certified Copy Attached? NO
199 21 209.0	Germany	05/07/1999	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>	<input checked="" type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>

Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
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DECLARATION**Page 2**

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP00/03852	04/28/2000	

Additional U.S. or PCT International application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

<input type="checkbox"/> Firm Name OR <input checked="" type="checkbox"/> List Attorney(s) and/or agent(s) name and registration number below:	Customer Number or label		
Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke Kimberly R. Hild	21,062 39,224	Glenn E. J. Murphy Stephen D. Harper	33,539 33,243

Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

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		Fax	610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:		<input type="checkbox"/> A petition has been filed for this unsigned				
Given Name	Juergen	Middle Initial	Family Name	LORENZ	Suffix e.g. Jr.	
Inventor's Signature			Den	Date	8.11.01	
Residence: City	Bopfingen	State	Country	Germany	Citizenship	Germany
Post Office Address		Brucknerstrasse 8				
Post Office Address						
City	73441 Bopfingen	State	Zip	Country	Germany	Applicant Authority
<input checked="" type="checkbox"/> Additional inventors are being named on supplemental sheet(s) attached hereto						

Type a plus sign (+) inside this box

H 4136 PCT/US

DECLARATION**ADDITIONAL INVENTOR(S)
Supplemental Sheet**

Name of Additional Joint Inventor, if any: A petition has been filed for this unsigned inventor

Given Name	Ewald	Middle Initial		Family Name	WILKA	Suffix e.g. Jr.	
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Inventor's Signature	<i>Ewald Wilka</i>				Date	26.11.03	
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Residence: City	Unterschneidheim	State		Country	Germany	Citizenship	Germany
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Post Office Address	Schillerstrasse 16 <i>Dex</i>						
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City	73485 Unterschneidheim	State		Zip		Country	Germany	Applicant Authority
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Name of Additional Joint Inventor, if any: A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name				Suffix e.g. Jr.	
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Inventor's Signature					Date			
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Name of Additional Joint Inventor, if any: A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name				Suffix e.g. Jr.	
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Inventor's Signature					Date			
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Name of Additional Joint Inventor, if any: A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name				Suffix e.g. Jr.	
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Inventor's Signature					Date			
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City		State		Zip		Country		Applicant Authority
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Additional inventors are being named on supplemental sheet(s) attached hereto